ROTATIONAL RELAXATION AND SELF-INDUCED TRANSPARENCY IN HF GAS

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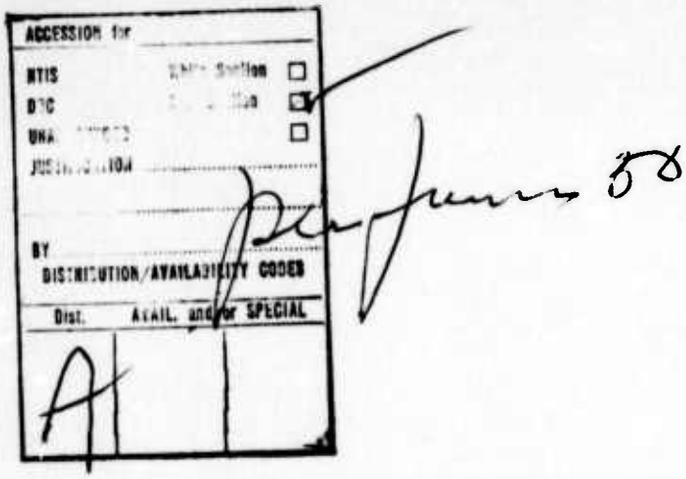

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20 ABSTRACT (Continue on reverse side if necessary and identify by block number)

Energy transfer rate measurements between J-states in vibrationally excited HF (hydrogen fluoride) gas were first attempted using a laser-excited infrared double-resonance technique. However, the pulsed HF "pin" lasers were found to be extremely narrow spectrally, making spectral coincidence extremely difficult. This led us to abandon the double-resonance approach.

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20. Abstract (continued)

One of the significant accomplishments reported is the measurement of laser linewidth using time-resolved bleachable absorption of HF gas. The pin lasers were found to have a width of only 3 MHz, which is 120 times narrower than the Doppler width of the lasing transition. This made the probability of obtaining spectral coincidence between the two HF lasers extremely remote.

In place of the two-laser double-resonance approach, we adopted a single-laser pump-probe technique whereby a single laser pulse of microsecond duration is broken into saturating pumping radiation followed by weak probing radiation using a Pockel's cell optical switch. Spectral coincidence (and beam alignment) is no longer a problem with this approach. Preliminary measurements of rotational relaxation times in HF at several tens of millitorr are presented for several $P_1(J)$ transitions; the values obtained are in agreement with values obtained from Lorentz pressure-broadening data.

At a high incident laser intensity, the absorbing transition of the HF was saturated (bleached) and the later portions of the laser pulse passed without attenuation. In addition to this bleaching, transmitted pulses were obtained whereby the energy absorbed from the leading edge appeared later in the pulse. In portions of the transmitted pulse the energy actually exceeded that of the incident pulse and the net energy loss was anomalously small. At the highest laser intensities, damped oscillations were observed in the transmitted output. These anomalies are believed the result of self-induced transparency and optical nutations, respectively. Nonlinear propagation may be valuable in obtaining lossless transmission through the absorbing atmosphere at $\lambda 2.7\mu m$, or for shaping HF laser pulses to attain short pulse widths and high peak power.

PREFACE

An investigation of fundamental molecular processes in support of laser system design was begun for the Advanced Research Projects Agency under Contracts Nr. DAAH01-72-C-0573 and Nr. DAHC-15-67-C-0062 at the Environmental Research Institute of Michigan (ERIM), formerly the Willow Run Laboratories of The University of Michigan. The effort currently reported, performed under Contract Nr. DAAH01-73-C-0591 and extending the earlier work, examines the transfer of rotational energy between HF molecules.

Director of the program at ERIM is R. R. Legault. G. H. Lindquist serves as Principal Investigator. The authors wish to thank Q. F. Carioti and R. A. Valade of ERIM for their able technical assistance.

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ROTATIONAL RELAXATION AND SELF-INDUCED TRANSPARENCY IN HF GAS

1
INTRODUCTION AND SUMMARY

The kinetics in high-powered chemical lasers are reasonably well understood, particularly concerning the partition of vibrational energy in the chemical reaction and the dissipation of vibrational energy through radiative and relaxation mechanisms. The success of laser performance predictions demonstrated by the RESALE program [1] and the successful construction of both large and small HF laser systems attest to this. However, the assumption invariably made in the design and analysis of such systems is that rotational relaxation occurs instantaneously—i.e., that the rotational energy in each vibrational state is always in equilibrium at some temperature, often the translational temperature of the gas. On the other hand, the rotational energy of the products created in reactions commonly used in HF lasers is known to be distributed in a very non-Boltzmann fashion: substantial fractions of the molecular products are produced in very high rotational states [2-5]. Therefore, before high-powered laser systems can be completely analyzed, it is necessary to determine at what rate rotational relaxation occurs and to show that a Boltzmann distribution of rotational averages is indeed produced after a given time.

The primary objective of this program has been to make direct measurements of rotational energy transfer, using a laser pumping-probing technique. A single rotational level of the sample gas is pumped from the first to the second vibrational level by means of a laser operating on the frequency of the P-branch fundamental vibration-rotation line of that rotational level. Sufficient power is used in the pumping beam to shift the population of the pumped state the

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maximum amount possible—i.e., to that condition in which the probabilities of stimulated absorption and stimulated emission are identical. The stimulated gas is then probed with a second beam at either the same frequency or a different one corresponding to either the pumped rotational level or to other rotational levels. By observing the action of the gas sample on the probing beam (usually absorption), the change in population of the state of the gas as a function of time and rotational level can be observed. Obviously, this system is only usable with either tunable lasers or with gas lasers which can be made to lase on their vibration-rotation transitions. The current interest is in HF and DF gas—both of which can be easily made to lase in their vibration-rotation bands using simple electric-discharge "pin" lasers.

The effort in this area on the previous contract was primarily spent in readying instrumentation [6]; the current contract was to have been primarily concerned with data acquisition on various gas mixtures. Initial data acquisition efforts were hindered, however, by a number of physical factors which had not been uncovered during the first year. These problems stemmed from the unexpectedly narrow spectral width of the laser pulse in conjunction with the transfer of translational energy (or lack of it) in the gas sample. Consequently, a very large part of the first half of this contract was spent in solving problems which, directly or indirectly, were a result of these factors.

Two parameters of the laser which were undetermined at the end of the last contract had profound effects on the operation of the experiment: (a) the spectral width of the laser line, and (b) the variation in laser frequency from pulse to pulse. Both are important because the pressure in the gas sample cell is low enough so that translational equilibrium cannot occur during laser excitation pulses. This means that the laser pulse bleaches a "hole" in the Doppler population profile of the lower state, and produces an upper-state population concentrated in the portion of the Doppler profile corresponding to the hole in the lower state (see Figure 1). If the probing of this stimulation process is to be successful, probe radiation must fall spectrally within the "hole." This condition dictates the degree of required frequency coincidence between the stimulating and probe beams.

One of the primary accomplishments during the first half of this contract was our determination of the spectral width and frequency stability of the HF lasers used to stimulate and probe the HF gas. The spectral width was measured by making carefully calibrated absorption measurements of laser pulses. These were then interpreted in conjunction with the Einstein coefficients [7] to yield spectral widths for the laser lines. The values were confirmed by ob-

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6. G. H. Lindquist, Investigations of Chemical Laser Processes, Final Report, 1 July 1972 to 31 December 1972, Report No. 191300-2-F, Environmental Research Institute of Michigan, Ann Arbor, June 1973.

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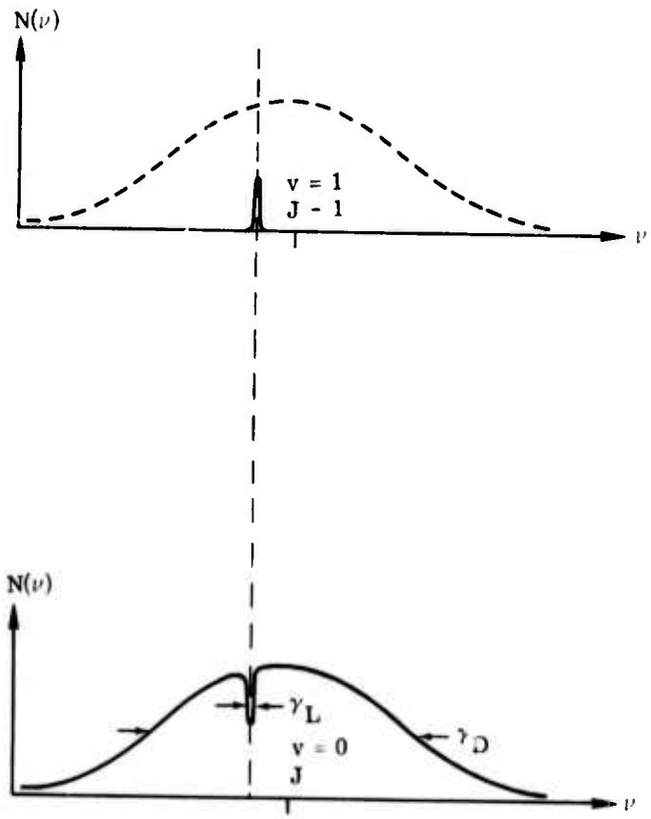

FIGURE 1. "HOLE" BURNED IN DOPPLER SPECTRAL DISTRIBUTION OF HF GAS MOLECULES BY INTENSE HF LASER PULSE. The $P_1(J)$ transition is shown; γ_D is the Doppler width and γ_L is the laser spectral width.

serving the variations in the heterodyne beating obtained in coincident beams from two similar HF lasers. This work is described in Section 2. The spectral width obtained is very much smaller than the best previous estimates [8]. It is approximately 0.0001 cm^{-1}, a factor of about 120 smaller than the Doppler width. This is the effective width of the "hole" produced in the Doppler distribution of the sample gas.

Furthermore, beating between coincident beams from two HF lasers, in particular the variations in the observed beat frequencies, yields a measure of the difference in frequency that can be expected in the operation of two supposedly identical HF lasers operating on the same spectral line. Frequency differences greater than 100 MHz (0.003 cm^{-1}) were observed. Since this is approximately the bandwidth limit of the measuring system, we assume that greater frequency differences could conceivably occur. The small spectral width compared to the frequency differences observed indicates that the chances of obtaining spectral coincidence of two lasers, and hence of having a probe laser operate within the frequency band corresponding to the "hole" produced by the stimulating laser, are small.

This frequency difference makes it very difficult, indeed, to perform a rotation-dependent relaxation experiment using two lasers; hence, for the time being we pursued relaxation measurements utilizing only one laser in which the same laser pulse serves as both stimulating beam and probe beam. In such a case, frequency coincidence problems do not occur. The first such effort involved a multiple-pass White cell as an optical delay line. After stimulating the gas in the sample cell, the stimulating pulse was delayed and returned through the sample cell to act as a probe pulse. But various problems occurred with this scheme. The long tails on our laser pulses and slight differences between the laser frequency and the peak of the Doppler profile made the experimental scheme difficult to utilize. Currently we are using a Pockel's cell, made of cadmium telluride, to switch off the stimulating pulse after the bleaching of the cell. That leakage radiation not extinguished by the crossed polarizer of the Pockel's switch serves as a probe pulse. The Pockel's cell has recently been completed; Section 3 describes some preliminary measurements made using this technique. The forthcoming final report will describe these and other measurements made during the latter half of the contract.

The relaxation times obtained agree reasonably well with those predicted from collisional linewidths in pure HF. This is to be expected since the collisions that are important in relaxing the particular type of stimulation produced are the same as those which produce collisional broadening of spectral absorption lines.

At the end of the previous contract, rotational relaxation was observed by noting the variation of absorbed laser power as a function of sample cell HF pressure. This technique made

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8. J. Goldhar, R. M. Osgood, Jr., and A. Javan, Observation of Intense Superradiant Emission in the High-Gain Infrared Transitions of HF and DF Molecules, Appl. Phys. Letters, 18, p. 167, (1971)

use of the following ideas. At very low pressure, bleaching in the sample gas, as well as the attendant variation in absorption, occurs because the molecules do not relax significantly during the time the pulse is on. As the pressure increases and relaxation processes (both translational and rotational) become important, the absorption goes to the usual equilibrium value given by the use of a Boltzmann distribution, unchanging with time. This variation in absorption with pressure and laser power can be used to obtain a relaxation time. Previous analysis of this type [6] did not yield usable results because no accurate values were available for either laser spectral width or laser pulse energy. Since these discrepancies were eliminated during the current contract, we were able to attempt such an analysis. In conjunction with this effort we have succeeded in obtaining a fully time-resolved laser pulse essentially free from electrical interference effects, whereas previously we had to be satisfied with measures of the integrated pulse. But these time-resolved traces are complicated by the appearance of coherent, induced emission. A coupling exists between the radiation field of the propagating pulse and the polarization of the resonant medium. Radiation absorbed by the medium is not lost but appears later in the pulse as induced emission. This self-induced transparency definitely contributes to the transmission of the medium and must be understood before an analysis of the variation of bleaching as a function of cell pressure and laser power is complete. Undoubtedly these effects were certainly present in the effort made under the previous contract, but they could not be observed because of the lack of time-resolution Such effects were probably the primary reason for the poor fit between theory and experiment in that analysis [6]. In view of the very large effect of self-induced transparency upon the bleaching of an HF cell, and the difficulty of including effects in a model for bleaching as a function of cell pressure and laser power, we have tentatively abandoned this approach to relaxation determinations. Section 4 briefly describes the physical process leading to these molecular coherence effects and presents some observations of them.

This report primarily covers the first six months of the contract, but as the prospective date of its publication was pushed back, partial results from later portions of the contract were included, especially when they represented significant advances.

2
PULSED HF CHEMICAL LASER LINEWIDTH MEASUREMENTS USING TIME-RESOLVED BLEACHABLE ABSORPTION OF HF GAS

The spectral linewidth of a high-gain, pulsed, infrared laser is difficult to measure because of (1) pulse-to-pulse shifts in laser frequency that preclude the use of scanning Fabry-Perot interferometry and (2) the absence of photographic sensitivity in the IR which prohibits the recording of single-pulse interferograms. Attempts to measure the linewidth of an HF chemical laser by Goldhar et al. [8], who used a pneumatically tuned Fabry-Perot interferometer, indicated that the spectrum was distributed (as averaged over several pulses) within a frequency

interval below the 70 MHz resolution limit of their instrument. We report in this section the measurement of the spectral linewidth at 2.7 microns of a pulsed HF chemical laser using a technique of saturable absorption of low pressure gas in an external cell. Our measurements, using the v = 0 to v = 1, P(J) transitions of HF gas, show that the laser linewidth is 0.0001 cm^{-1} or 3 MHz (which is about 120 times narrower than the Doppler spectral width) and approaches the Heisenberg uncertainty limit established by the laser pulse duration.

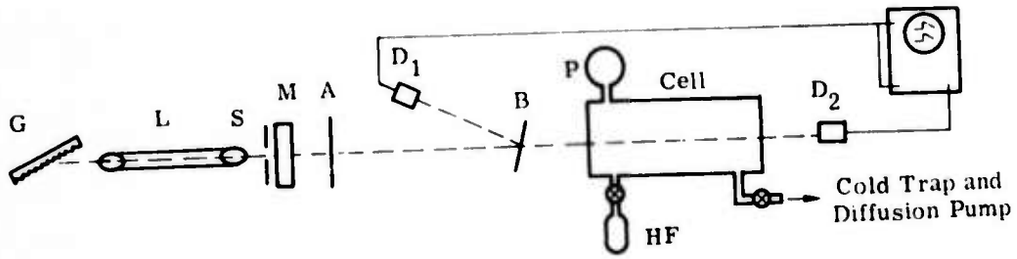
The experimental apparatus used is illustrated in Figure 2. A 0.2 m helical "pin" laser, L, was used as a source. Single-line power output was on the order of 10 W. Excitation was provided by four 500 pF "door knob" capacitors in parallel charged to 20 kV and switched with a hydrogen thyratron. Single-line operation was achieved with a 625 ℓ/mm grating, G, and output coupling was obtained with an uncoated flat germanium mirror, M, with a transmission of 50%. The 0.3 m HF absorption cell, made of Monel, was heated to 100°C to eliminate HF dimers. The HF pressure was maintained in a constant flow configuration* and monitored with a capacitance manometer, P, with a pressure range of 1 to 10^3 mTorr. A Monel HF-supply cylinder was maintained at -85°C in order to freeze out impurities (particularly water) and to reduce the HF vapor pressure so that a desirable gas throughput could be achieved.

Two 0.02 mm^2 InAs detectors, D_1 and D_2, monitored the temporal profile of the laser pulses before and after the absorption cell. They were back-biased to achieve maximum frequency response and their outputs were displayed on a dual-beam oscilloscope having a measured 2.4 nsec response time. The arrival of a signal from detector D_1 provided the oscilloscope trigger for both traces. The detectors were calibrated using a 1000°C blackbody and a $\lambda 2.7 \mu m$ filter, enabling direct measurements of the peak power density of the laser beam.

Figure 3 shows representative transmitted laser pulses for HF absorption cell pressures of 0, 10, 20, and 40 mTorr. Each waveform is a superposition of about ten pulses and shows good reproducibility. The waveform at zero pressure is of course the unattenuated incident laser pulse. Introduction of HF gas into the cell attenuates the leading edge of the incident pulse according to Beer's law at equilibrium. This absorption substantially populates the upper state of a vibration-rotation transition such that the optical transmission at the laser frequency increases rapidly with time. When the upper-state population approaches that of the lower state, the cell is "bleached," and the later portion of the laser pulse is transmitted with little attenuation.

Since the spectral width of the laser pulse is less than the Doppler width of the absorbing HF gas, a "hole" equal to the laser linewidth is "burned" in the Doppler profile. Simple measurement of the amount of laser energy absorbed at a particular gas pressure allows one to calculate the number of molecules excited and thereby determine the laser spectral linewidth.

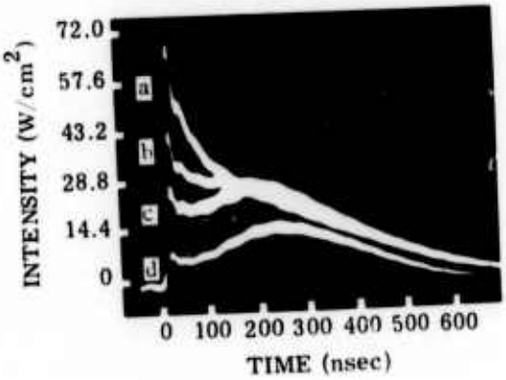
*Static, nonflowing HF gave identical results.



KEY

G = grating
L = 0.2m "pin" laser
S = 1.5mm aperture
M = output mirror
A = calibrated neutral density attenuator
B = beam splitter
D_1 and D_2 = InAs detectors
P = capacitance manometer

FIGURE 2. EXPERIMENTAL SETUP FOR BLEACHING EXPERIMENT TO DETERMINE LASER LINEWIDTHS



KEY

HF Gas Pressure (mTorr)
(a) 0
(b) 10
(c) 20
(d) 40

FIGURE 3. TRANSMITTED $P_1(5)$ HF LASER LINES FOR SEVERAL ABSORPTION CELL PRESSURES

It should be noted that if the intensity is too high, the laser pulse can bleach a "hole" wider than the laser linewidth in the Doppler spectrum. In our experiment, however, care was taken to control the incident laser intensity by placing neutral density attenuators in the beam such that the incident intensity was low, yet sufficient to bleach the cell. Moreover, if a wider "hole" were produced, the width would decrease with increasing gas pressure. This is clearly not the case for our experiment; we feel that our values are a good representation of the laser linewidth and not simply an upper limit.

For this technique to be a true measure of the laser linewidth, no appreciable molecular relaxation back to equilibrium can occur in either the lower or the upper vibration-rotation state. This is certainly true for gas pressures in the mTorr range. The most rapid relaxation process is rotational relaxation caused by long-range multipole interactions which, from collisional linewidth data [9], has a relaxation rate on the order of 10^8 sec^{-1} $Torr^{-1}$. The short laser pulse durations used in this experiment allowed us to ignore rotational relaxation at low gas pressures. Translational [10] and vibrational [11] relaxation rates are one and three orders of magnitude slower, respectively, than that of rotation and may clearly be ignored.

The transmitted pulses of Figure 3 show that for the lower HF pressures little relaxation is present. Once bleaching has occurred, very little laser energy is consumed in maintaining the bleached condition. At higher pressures, however, there are sufficient collisions to induce rotational transitions in both the lower and upper vibrational states, requiring additional photon absorption to maintain the bleached condition.

Table 1 presents the linewidth and absorption measurements of four $P_1(J)$ lines*. The HF laser was operated with an H_2:SF_6 pressure ratio of 10:20 Torr and was pumped at 18 kV. The equilibrium absorption coefficient, α, was calculated using

$$\alpha = \frac{B(0,J;\,1,J-1)}{c\gamma_D/2}\left(\frac{\ell n 2}{\pi}\right)^{1/2} N(2J+1)\exp\left[-\frac{hcB_0}{kT}J(J+1)\right] \qquad (1)$$

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*The notation $P_1(J)$ refers to P-branch lasing transitions where $(v = 1, J - 1) \rightarrow (v = 0, J)$.

TABLE 1. LASER LINEWIDTH MEASUREMENTS

Laser Transition	Absorption Coefficient of HF Gas (cm^{-1} $Torr^{-1}$)		Peak Laser Intensity (W/cm^2)	Pulse Duration (nsec)	Laser Linewidth (cm^{-1})
	Calculated	Measured			
$P_1(3)$	3.411	3.54	183	55	0.000 168(±7)
$P_1(4)$	2.535	2.59	130	110	0.000 094(±7)
$P_1(5)$	1.506	1.52	128	210	0.000 112(±4)
$P_1(6)$	0.732	0.71	43	260	0.000 101(±10)

where $B(0,J;\ 1,J-1)$ is the Einstein B for the $P_1(J)$ transition

γ_D is the Doppler full-width at half-height

J is the rotational quantum number of the lower state

T is the gas temperature

B_0 is the rotational constant for the v = 0 vibrational level

The $B(v,J;\ v',J')$ were determined using the Einstein A values [7] and $B = A/8\pi\tilde{\nu}^2$. The leading edge of the bleaching laser pulses, as well as the entire pulse of suitably attenuated nonbleaching laser pulses, experienced absorption in agreement with the calculated values (as demonstrated in the table).

The laser linewidth, γ_L, was calculated according to the following:

$$N_a = \frac{1}{2}\left(\frac{2J-1}{2J+1}\right) N(\tilde{\nu})\gamma_L \tag{2}$$

where N_a is the number density of photons absorbed and, therefore, the number of molecules excited per unit volume, and $N(\tilde{\nu})$ is the molecular number density per frequency interval at the peak of the Doppler distribution for the v = 0, J-state. The degenerate m_J-states in a J-level are not equally populated during saturation since only transitions which satisfy the selection rule $\Delta m_J = 0, \pm 1$ can participate in the absorption, and orientational relaxation between adjacent m_J-states is an order of magnitude slower than rotational relaxation between adjacent J-levels. Thus, the factor $(1/2)[(2J-1)/(2J+1)]$ takes into account the difference in degeneracy between the upper and lower vibration-rotation levels and includes only those transitions allowed according to the selection rule for m_J. $N(\tilde{\nu})$ may be determined from the Doppler spectral profile and the partition of energy distributed to each rotational state. We then have

$$\gamma_L = \frac{4J+2}{2J-1}\left(\frac{E_a}{h\tilde{\nu}c}\right)\left\{\frac{2}{\gamma_D}\left(\frac{\ln 2}{\pi}\right)^{1/2} N(2J+1)\frac{hcB_0}{kT}\exp\left[-\frac{hcB_0}{kT}J(J+1)\right]\right\} \tag{3}$$

where E_a is the amount of laser energy absorbed per cm^3.

Because the calculation of γ_L is almost identical to that of α, and since agreement between our theoretical and observed values of the absorption coefficient is better than 4%, one expects similar accuracy for the linewidth values (assuming an accurate determination of E_a). Each linewidth value in the table is an average taken over three different absorption pressures; the standard deviation is indicated in parentheses. Although the laser output intensity and pulse duration varied appreciably from line to line, there was little change in the spectral linewidth. Less attenuation was used for the $P_1(3)$ line measurements, and excessive "hole-burning" may account for the larger spectral width measured.

By decreasing the H_2 pressure in the laser discharge, it was possible to increase the laser pulse duration of a given line by a factor of 10. The laser linewidth, however, decreased by only a factor of 2.

The variation in the frequency of the laser pulse was examined by detecting the heterodyne beating between two HF laser beams. Figure 4 shows the beating observed and the relative frequency shift from pulse to pulse. The frequency of each P(J) line could be grossly adjusted by fine-tuning the grating, and the lowest beat notes were obtained by fine adjustment of the grating tilt. Beats with periods greater than the pulse duration could be obtained for a few seconds but would then drift to higher frequency, presumably as the result of small temperature changes or mechanical disturbances. It should be noted that the beating is uniform, indicating no appreciable change in frequency within the time duration of the pulse.

The near field and far field (at the focus of a 50cm lens) patterns of the laser output were scanned with the $0.02mm^2$ InAs detector. The laser beam was found to be uniform and its divergence was determined to be 2.2 mrad—equal to the divergence limit of the 1.5mm limiting aperture of the laser. Scanning of the coincident beams during heterodyne beating was also performed. No appreciable change in beat frequency was observed as the detector position was changed, indicating spectral uniformity of the laser output beams.

Interesting observations were made at high laser outputs and low HF pressures. Transmitted laser pulses could be obtained whereby the energy absorbed from the leading edge appeared later in the pulse. The optical intensity in portions of the transmitted pulse actually exceeded that of the incident pulse, and the net energy loss was anomalously small. When the difference between the incident and transmitted pulses was displayed on an oscilloscope, we obtained a damped ringing trace composed of absorption followed by emission identical to optical nutations [12]. This anomalous transmission is believed to be self-induced transparency [13]; the results of this study are described in Section 4.

3

RELAXATION MEASUREMENTS IN HF GAS USING PULSED LASER SATURATION

3.1 INTRODUCTION

The primary objective of these studies is to determine experimentally the populations of HF molecules as a function of time and rotational quantum number J. Pulsed laser saturation spectroscopy was used, whereby intense pumping radiation from a particular HF laser transition saturated the absorption of HF in an external cell. The transition was then probed as a function of time to determine the relaxation rate back to equilibrium.

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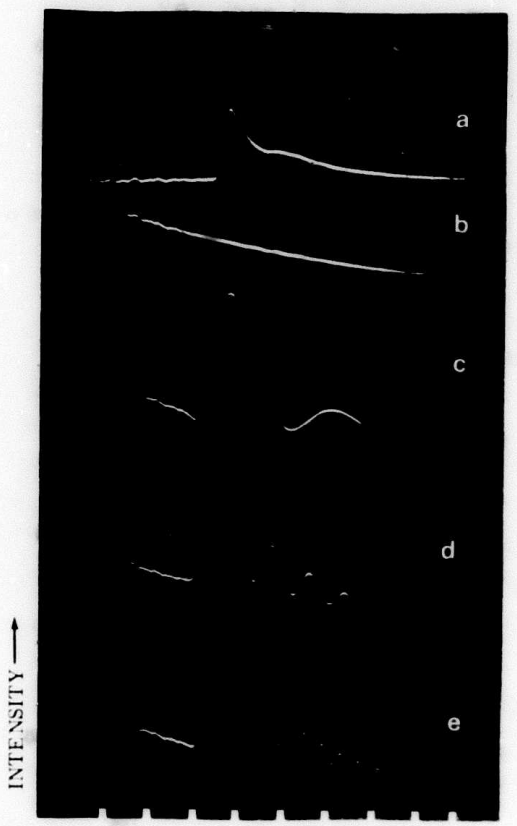


FIGURE 4. HETERODYNE BEATING OF TWO HF LASERS

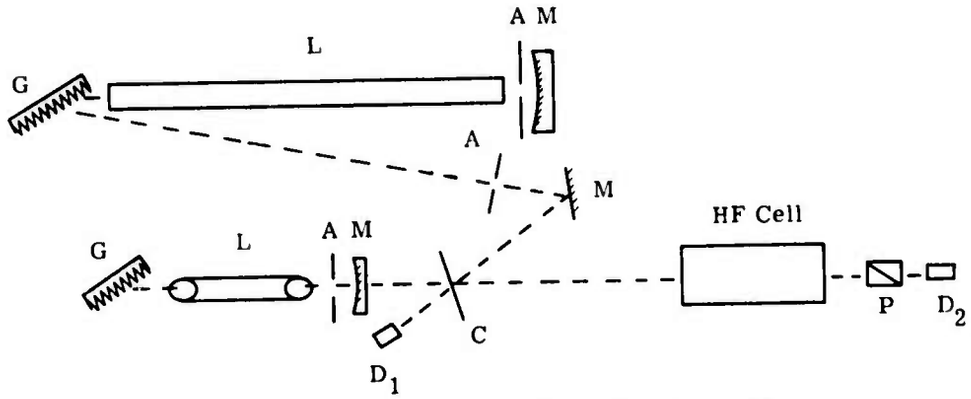
A two-laser, infrared double-resonance approach was attempted using a $P_1(J)$ line for pumping and a $P_2(J')$ line for probing. By probing at several J', both the redistribution of energy from the initially excited J-state and the respective redistribution rates could be determined. Figure 5(a) illustrates the experimental arrangement. Unfortunately, the laser spectral width was found to be extremely narrow — 120 times narrower than the Doppler width (see Section 2). The width was narrower than the pulse-to-pulse frequency changes of the laser and prevented the probe from interrogating the narrow region of excitation within the Doppler distribution. Even when the probe was operated with zero delay time on the same $P_1(J)$ transition as the pump, it was unaffected by the presence of the saturating pump radiation.

Figure 5(b) illustrates the second approach for measuring relaxation time. Here, frequency coincidence was no problem since pump and probe radiation came from the same pulse. A White cell adjustable for multiple passes provided the variable delay time for the probing radiation which was split off from the pumping pulse. We observed, as expected, that the pumping pulse "bleached a hole" through the gas cell for the probing radiation, but the long tail of the excitation pulse prevented us from observing any significant relaxation from the saturated state.

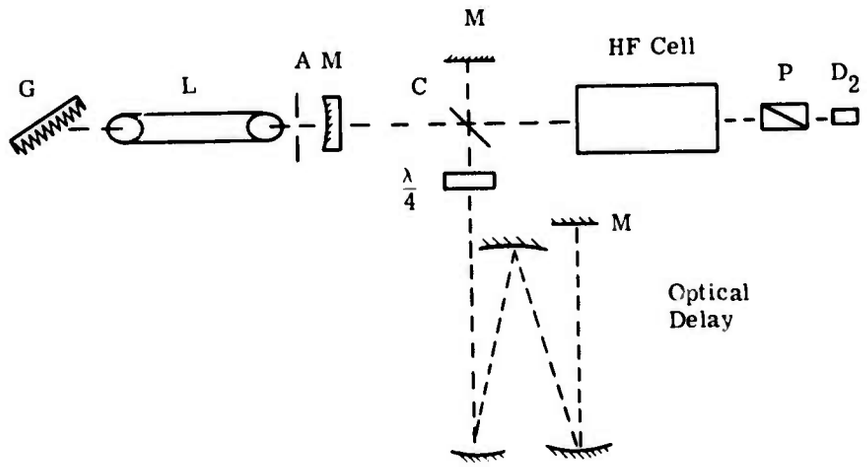
Our most recent attempt at relaxation measurements also involved a single pulse split into both pump and probe ratiation. Here, however, a Pockel's cell allowed the initial portion of the pulse to saturate the transition; then it abruptly turned the pulse off, allowing only a small fraction of the radiation through to provide the probe. Preliminary measurements have been completed, and the relaxation times obtained are in agreement with those expected from theoretical considerations.

3.2 EXPERIMENTAL ARRANGEMENT

Figure 6 illustrates the experimental arrangement using a Pockel's cell to split a single laser pulse into pumping and probing radiation. Vertically polarized radiation from the HF laser initially passes through the Pockel's cell (zero applied voltage), is unimpeded by the Glann-Thompson polarizer, P, double-passes the HF gas cell to saturate the absorbing transition, and returns to the germanium reflector, Ge, oriented at Brewster's angle so that very little of this intense vertically polarized pumping radiation reaches the detector, D_2. After about 200 nsec of the 1 μsec pulse has passed, the Pockel's cell receives a fast-rise-time, high-voltage signal which rotates the polarization of the radiation by 180^0. Only a small amount of vertically polarized light is passed by the polarizer, P, and this serves as the weak intensity probe of the excited HF gas. The probe intensity is less than that of the pump by about two orders of magnitude. This weak, vertically polarized radiation also double-passes the HF gas cell, is passed by P, and undergoes a 180^0 rotation in polarization by the Pockel's cell, P.C., whose high voltage is still on. The polarization is now horizontal and is selectively reflected, by the Ge, to the detector. It was necessary to double-pass the Pockel's cell in order to prevent intense pumping radiation from saturating the detector and distorting the probing signal. This experimental scheme had the advantage of providing a long, roughly 800 nsec probe so that a single pulse provided an entire range of delay times.



(a) Two-Laser, Pump-Delay-Probe Approach

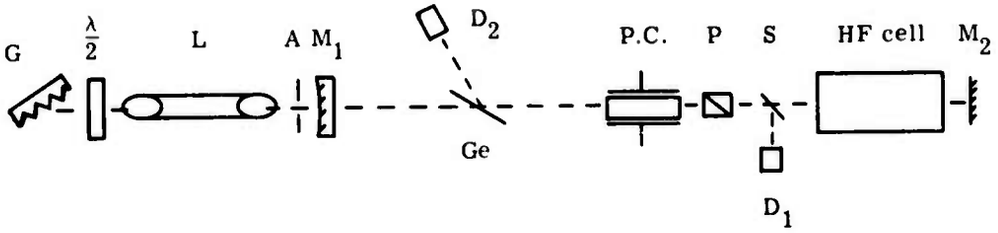


(b) Single-Laser Approach in Which Part of the Pump Is Optically Delayed and Returned to Probe the Gas

KEY

G = grating
L = transverse-discharge helical "pin" laser
A = aperture
M = mirror
C = beam combiner
D_1 and D_2 = InAs detectors
P = Glann-Thompson polarizer
$\lambda/4$ = sapphire quarter-wave plate

FIGURE 5. TWO EXPERIMENTAL SCHEMES FOR ATTEMPTING RELAXATION MEASUREMENTS



KEY

G = grating
$\frac{\lambda}{2}$ = sapphire half-wave plate
L = transverse-discharge helical "pin" laser
A = aperture
M_1 and M_2 = mirrors
Ge = germanium Brewster reflector
P.C. = Pockel's cell for optical switching
P = Glann-Thompson polarizer (oriented parallel to laser polarization)
S = glass beam splitter
D_1 and D_2 = InAs detectors

FIGURE 6. PULSED LASER SATURATION EXPERIMENT USING A SINGLE PULSE AS BOTH PUMP AND PROBE

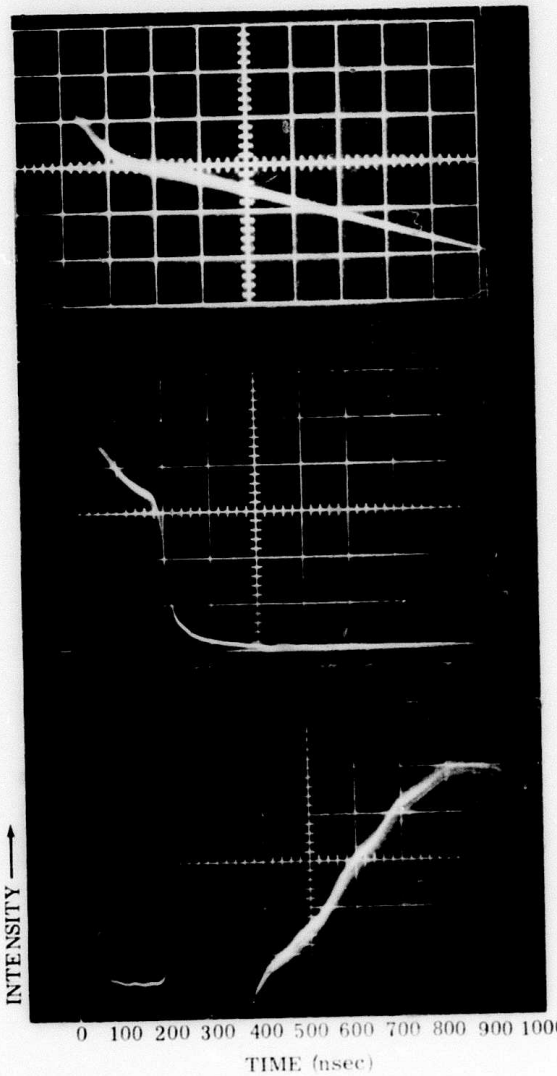
The parts of Figure 7 show the following: (a) the output from the HF laser, (b) the optically switched pulse which pumps and probes the HF cell, and (c) at a scale expanded 400-fold, the probing signal as obtained by the final detector D_2. Detection of the weak probe signal is initially affected by the switching of the Pockel's cell but recovers again within 200 nsec. A high voltage pulse having a fast rise-time and controllable, exponential fall-time, when applied to the Pockel's cell, produces the probe waveforms of Figure 7(c).

3.3 EXPERIMENTAL RESULTS

Introduction of HF gas into the absorption cell attenuated the leading edge of the pumping pulse as the gas was excited into saturation. The transmission of the gas to the probing radiation was initially large, due to saturation of the transition. The probe transmission decreased with time, however, as the population of the excited state was redistributed. When transmission of the probing radiation was plotted as a function of time, an exponential curve was obtained whose time constant was a measure of the fastest relaxation mechanism—principally rotational relaxation resulting from long range collisions involving changes in angular-momentum quantum number J.

Table 2 shows our preliminary data on three lasing transitions at several HF gas pressures. In Figure 8 these data are plotted and compared with relaxation times expected from collisional linewidth data at near atmospheric pressures [9]. A relaxation time obtained from the decay rate of an optical nutation in HF gas is also plotted (for a discussion of this effect, see Section 4).

The results obtained agree reasonably well with collisional linewidth measurements, although the relaxation is somewhat slower than linewidths would indicate. In this experiment, the relaxation mechanism will be that process which most rapidly fills the hole in the $v = 0$, J-state and flattens the population peak produced in the $v = 1$, $(J - 1)$-state. The departure from equilibrium produced by the laser stimulation is very small, in that the fraction of molecules excited is small and only two vibration-rotation states are involved. The relaxation rate will essentially correspond to the rate at which excited molecules leave the upper state of the stimulating transition or, equivalently, enter the lower state of the transition. (Because of the small departure from equilibrium, such rates are practically the same as corresponding rates for HF gas at equilibrium.) These rates are important in collisional broadening of spectral lines. In particular, the collisional width of a spectral line is a measure of the mean time that a single molecule remains in either the upper or lower state of a transition (i.e., the time between collisions which take it into or out of the two states); this time, in turn, is the same as the relaxation time of a small departure from equilibrium in such a system. Thus, since collisional linewidth data can indicate something about rotational relaxation rates under certain specific stimulation conditions, the agreement of our preliminary measurements with such data is to be expected.



(a) $P_1(5)$ HF Laser Output

(b) Optically "switched" Laser Output (strong pumping radiation is followed by weak probing radiation — finite detector response limits the fall-time at switching)

(c) Extinguished Pump Followed by Probing Signal. Vertical scale is 400 × that in (a) and (b)

FIGURE 7. OSCILLOSCOPE TRACES OF PUMPING AND PROBING RADIATION

TABLE 2. HF GAS RELAXATION TIME MEASUREMENTS

Lasing Transitions	HF Gas Cell Pressure (mTorr)	Relaxation Time (μsec)	τp (μsec-mTorr) Observed	Calculated
$P_1(5)$	21.4	0.65	14	13
	36.0	0.60	22	
	48.5	0.29	14	
*$P_1(5)$	113.0	0.14	15	13
$P_1(6)$	54.7	0.44	24	22
	66.0	0.42	28	
	106.0	0.34	36	
$P_1(7)$	108.0	0.42	46	37

*Data taken from the decay rate of an optical nutation in HF gas (see Section 4.3.2 of this report).

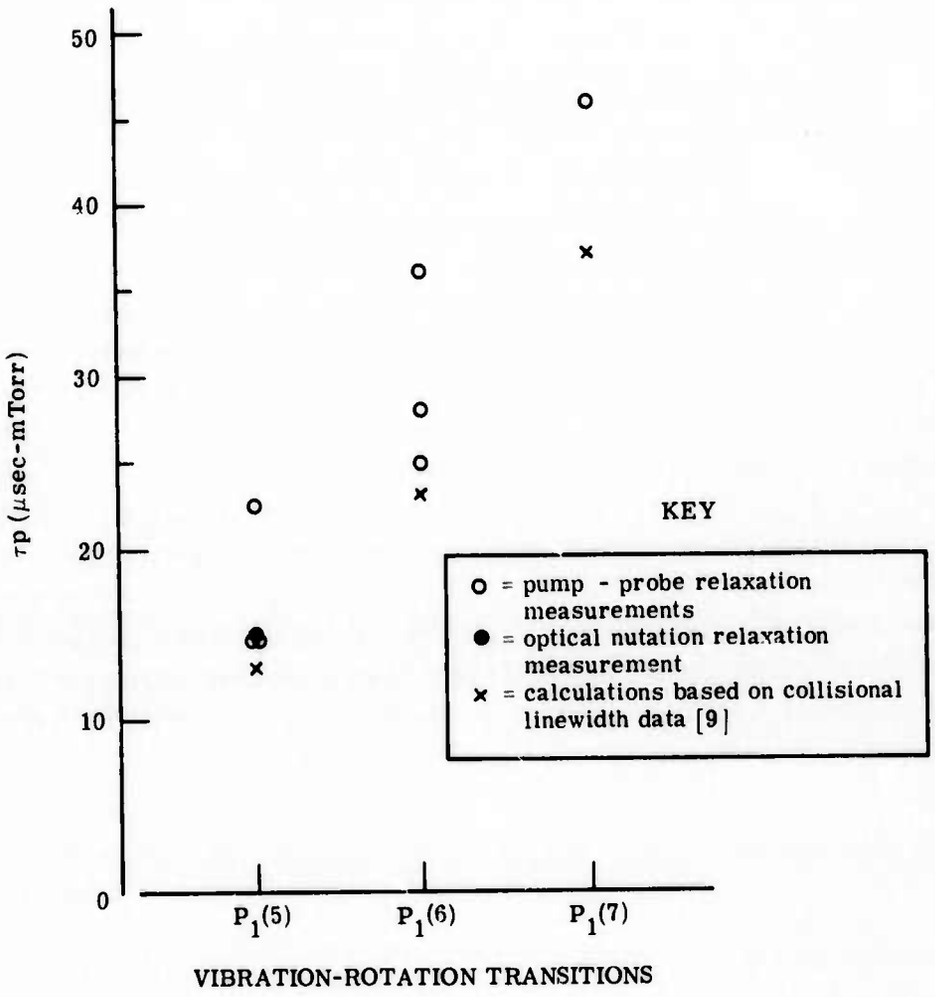


FIGURE 8. RELAXATION TIME FOR SEVERAL $P_1(J)$ TRANSITIONS

Agreement between our data taken at mTorr pressures and collisional linewidth data taken at near-atmospherie pressures verifies the expected linearity of relaxation rate with pressure. Moreover, collisional linewidth measurements require many absorption measurements at very high speetral resolution and have been performed for very few diluent gases. The experimental teehniques used here will easily yield results on any number of diluent gases and have the advantage of operating at low pressure where behavior is elose to ideal. We plan to take measurements using diluent gases and present them in the final report.

4

PULSE PROPAGATION IN RESONANT ABSORBERS: SELF-INDUCED TRANSPARENCY IN HF GAS

4.1 INTRODUCTION

Self-Indueed Transpareney (SIT) through an optieally resonant medium was first predieted and observed in a ruby erystal by McCall and Hahn [13] using ruby laser pulses; it has sinee been observed in SF_6 [14], CO_2 [15], and NH_3 [16] gases using infrared laser radiation at 10.6μ. Our laboratory has reeently observed self-indueed transparency and the related optieal nutation effeet [12] in HF gas using a transverse-diseharge, pulsed HF ehemieal infrared laser at λ 2.7μm. Since HF laser radiation is also optieally resonant with water vapor, SIT should be observable in water vapor—whieh offers an exeellent possibility for low-loss propagation through the atmosphere. In addition, SIT ean produee optical pulse eompression [17] and inereases in peak power. This method of pulse eompression to attain greater peak power eould be valuable in the use of HF ehemical lasers as powerful sourees of infrared radiation. In the following seetion, the theory of the self-indueed transpareney effect is deseribed briefly, along with some of its manifestations. Seetion 4.3 describes the observed results demonstrating self-indueed transpareney effeets. Seetion 4.4 contains some reeommendations for further study into the effects of self-indueed transpareney and its possible uses.

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4.2 THEORY

4.2.1 SELF-INDUCED TRANSPARENCY

Self-induced transparency is a nonlinear optical propagation effect occurring in media which are optically resonant with intense propagating radiation. It is observed when the radiation pulse duration is less than the relaxation time associated with the homogeneous linewidth of the medium and the power density of incident laser radiation is above a threshold value. The leading edge of the incident radiation is absorbed according to Beer's law but is re-emitted into the later portion of the pulse. Only de-phasing collisions occurring during the laser pulse produce attenuation. The majority of the total energy passes with very low loss.

A brief description of SIT follows. Referring to Figure 9, consider circularly polarized radiation of amplitude, E, and frequency, ω, propagating in the z-direction through the medium of interest. To simplify the discussion, we choose a frame of reference which is rotating with the E-vector at a frequency of ω. In this reference frame the macroscopic electric dipole, p, precesses at a frequency $\Omega = 2pE/\hbar$ about the E-field. Because of inhomogeneous broadening (for gases, Doppler broadening), the resonant frequency can differ slightly from ω. As time progresses, the individual dipoles de-phase and the macroscopic dipole fans out as a result of the distribution of Doppler frequencies. Two of these frequencies, ω^+ and ω^-, represent molecules whose resonance frequencies are respectively larger and smaller than ω. In our rotating frame of angular frequency ω, the dipole of frequency ω^+ precesses slowly about the $\hat{z}$-axis in a right-hand sense, while the dipole of ω^- precesses in the opposite sense. The de-phasing process continues for the first half-cycle of precession, but for the second half-cycle the direction of slow precession about the $\hat{z}$-axis is such that the dipoles are re-phased. When all frequency components are considered, the macroscopic dipole fans out as the individual dipoles are de-phased. Then during the second half-cycle, the dipoles are re-phased and the macroscopic dipole reformed so that at full cycle the dipoles all oscillate in phase to produce a burst of radiation.

The E-field described above is of the proper intensity and duration to provide full-cycle precession for a particular dipole transition. The radiation pulse that provides this E-field is called a 2π pulse and satisfies the relationship

$$\frac{2p}{\hbar}\int_0^\infty E(t)\,dt = 2\pi \qquad (4)$$

For a particular dipole transition and pulse duration, this equation establishes a threshold E-field and, thus, the incident intensity for observing self-induced transparency.

The above is a simple, intuitive argument for SIT. Rigorous theoretical considerations and detailed descriptions appear in the references. Also, when degeneracy of the energy levels involved in the transitions is included, Eq. (4) becomes (see [12])

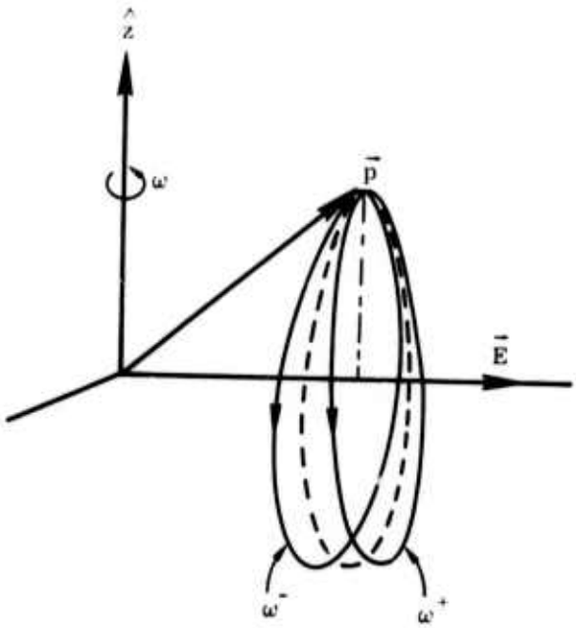


FIGURE 9. DIPOLE PRECESSION IN A FRAME ROTATING AT A FREQUENCY, ω, ABOUT THE $\hat{z}$-AXIS. Two Doppler components ω^+ and ω^- of the dipole $\vec{p}$ are chosen to illustrate the dephasing and rephasing effects in SIT. As the dipole components precess about the radiation $\vec{E}$-field, they also precess (due to frequency mismatch) about the $\hat{z}$-axis but with opposite senses. If $\omega^{\pm} = \omega$, the precession is about $\vec{E}$ only and not about the $\hat{z}$-axis.

$$\left[\frac{J_n}{6}\left(\frac{1}{2J_m+1}+\frac{1}{2J_n-1}\right)\right]^{1/2}\frac{p}{\hbar}\int_0^{\infty}E(t)\,dt = 2\pi \qquad (5)$$

where J_m and J_n refer to the angular momentum quantum numbers of the upper and lower states of the transition.

4.2.2 OPTICAL NUTATION EFFECT

If the E-field of the incident radiation is sufficiently strong, the dipoles may precess through several cycles (i.e., replacing 2π in Eq. (4) by $n\pi$ where n is even). The resulting oscillations in the transmitted radiation intensity are called optical nutations [12]—after nuclear transient nutations [18]. The oscillation frequency, Ω, for a $P_1(J)$ transition is

$$\Omega = \left[\frac{J}{6}\left(\frac{1}{2J+1}+\frac{1}{2J-1}\right)\right]^{1/2}\frac{|\bar{E}|\,p}{\hbar} \qquad (6)$$

where $|\bar{E}|$ is the average E-field amplitude—and again the terms under the radical take into account the degeneracy in the J-level [12].

4.2.3 PULSE COMPRESSION

An $n\pi$ pulse propagating through an absorbing medium will experience attenuation as a result of the inhomogeneous broadening mechanism if $n < 2$. Also, if $2 < n < 4$, it will be attenuated. In particular, if $n = 3$, absorption will be maximum and the 3π pulse will evolve into a 2π pulse as it propagates [17]. Consider now a 3π laser pulse focused into a medium so that, as the pulse propagates, SIT reshapes the 3π pulse into a 2π pulse but the focusing effect increases the energy density (and thus the E-field) to reshape the pulse back toward a 3π pulse. The result is a pulse which increases in peak intensity and decreases in temporal duration. Figure 10 shows the results of such an experiment by Gibbs and Slusher [17] using a 10-nsec HgII laser pulse whose radiation was focused into a resonant ^{87}Rb absorber. The pulse was compressed by a factor of 10 and the intensity increased by a factor of 3 or more.

4.2.4 LOSS RESULTING FROM HOMOGENEOUS RELAXATION

By ignoring homogeneous broadening due to collisions, theory shows that absorption of a 2π pulse is not only small but precisely zero [13]. When homogeneous broadening is included, however, the de-phasing effects (unlike those for inhomogeneous broadening) are irreversible and cannot be re-phased. These de-phasing collisions, which occur as the dipoles precess

18. A. Abragam, Principles of Nuclear Magnetism, Oxford Univ. Press, Cambridge, 1961.

about E, introduce loss to the system and reduce the magnitude of the "burst of radiation" when the macroscopic dipole is re-phased. However, if the incident laser pulse duration is short compared to the homogeneous relaxation time, the losses will be small.

Observation of the optical nutation effect provides a means of measuring the homogeneous relaxation time. Since the oscillations in the transmitted intensity decay according to this relaxation mechanism, the envelope of the oscillations provides a decay rate and, thus, a collisional relaxation time for the medium.

4.3 EXPERIMENTAL RESULTS

During the investigation of relaxation rates using pulsed laser saturation of low pressure HF gas (see Section 3 of this report), several interesting nonlinear propagation effects were observed.

4.3.1 OBSERVATION OF SELF-INDUCED TRANSPARENCY IN HF GAS

In using the experimental apparatus discussed and illustrated in Section 3, we observed anomalies in radiation transmission through HF which are consistent with those of self-induced transparency [13]. Figure 11 is an oscillogram depicting SIT of $P_1(5)$ radiation through HF gas in the 30cm sample cell. Each trace is the superposition of about ten laser pulses, and the four traces represent HF absorption cell pressures of 0 (i.e., the incident laser pulse itself), 26, 50, and 100 mTorr. At these pressures, the Beer's law transmittance for low intensity radiation at line center is 1.0, 0.31, 0.10, and 0.01, respectively.

SIT was observed for all the strong $P_1(J)$ lines of our laser (J = 3 - 7). Threshold intensities for SIT were in agreement with Eq. (5). For example, the effect did not occur below a power density of about 20 W/cm^2 when operating on the $P_1(5)$ line with a pulse duration of 350 nsec. The calculated power density for the SIT threshold is 23 W/cm^2, in agreement with our observed value.

Figure 12 illustrates the low loss as a result of SIT. Trace (a) shows the incident laser pulse and trace (b) displays the difference between the incident and transmitted intensities ($I_{out} - I_{in}$) for an HF gas pressure of 27 mTorr; the vertical scale in (b) has been expanded 2-1/2 times. The initial absorption appears later in the pulse as emission, and the amount of emission is almost identical to that absorbed, as may be seen by comparing the trace area below the baseline to that above. On the basis of Beer's law for weak intensity absorption, one would expect 70% absorption, yet the net absorption shown in Figure 12(b) is only a few percent.

4.3.2 OBSERVATION OF OPTICAL NUTATIONS IN HF GAS

When full power of our HF laser was propagated through the HF absorption cell, the optical nutation effect was observed. By operating the HF laser with a mixture lean in H_2 gas but with the usual amounts of SF_6—typically H_2:SF_6 = 1:20 Torr—the laser pulse duration was

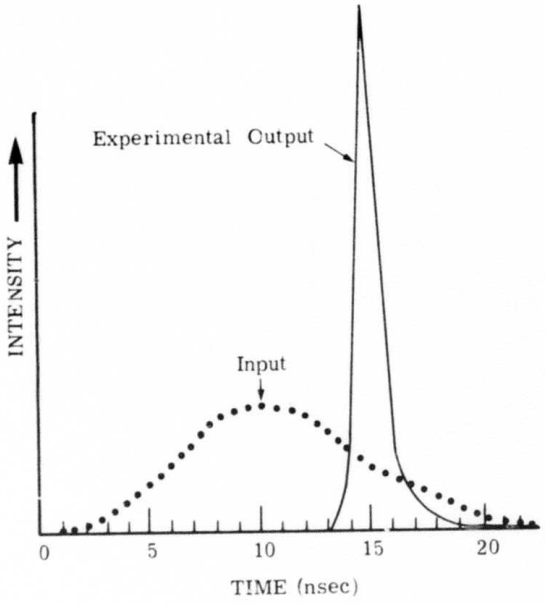


FIGURE 10. OPTICAL PULSE COMPRESSION AS OBSERVED BY GIBBS AND SLUSHER [17]

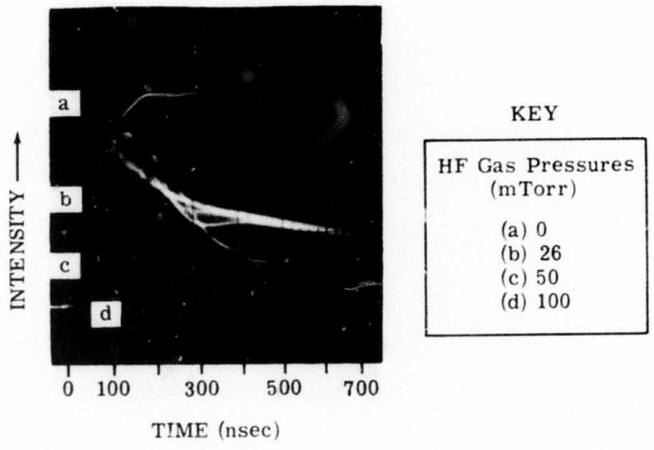
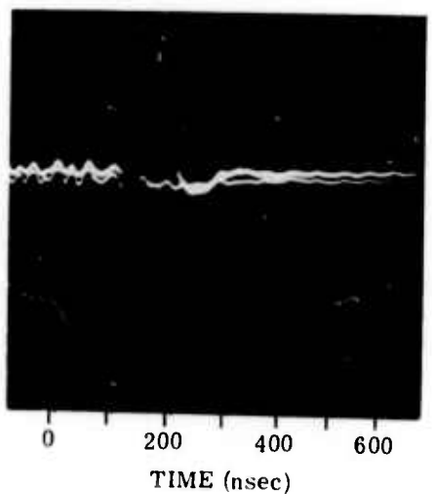


FIGURE 11. SELF-INDUCED TRANSPARENCY IN HF GAS AT SEVERAL PRESSURES FOR THE $P_1(5)$ HF LASER TRANSITION. The intensity of the transmitted pulses can actually exceed that of the incident radiation (a). Each trace is a superposition of about 10 pulses.

(a) Incident $P_1(5)$ Laser Pulse



(b) The Difference Between the Incident Pulse and That Transmitted Through 27 mTorr of HF Gas

FIGURE 12. LOW-LOSS PROPAGATION THROUGH ABSORBING HF. Note that initial absorption from the leading edge appears later in the pulse; the absorption is followed by almost complete emission. The vertical scale in (b) is 2.5× that in (a).

increased to approximately 1 μsec. With this long temporal pulse, the optical nutation effect was easily observed. Figure 13 shows a typical oscilloscope trace of incident and transmitted $P_1(5)$ pulses for an HF pressure of 113 mTorr—the weak intensity limit is less than 1% transmission.

The nutation frequency, Ω, is in good agreement with theory. The incident pulse in Figure 13 has an intensity of 88.5 W/cm^2 (E = 258 V/cm = 0.86 statV/cm), and the electric dipole matrix element, p, for the $P_1(5)$ transition is 11.19×10^{-20} esu-cm [7, 9]. Substitution into Eq. (6) yields $\Omega/2\pi$ = 6.9 MHz, which agrees well with the 140 nsec period or 7.1 MHz nutation frequency taken from the oscilloscope trace.

The relaxation time of the optical nutation was determined by plotting the transmission as a function of time and determining the exponential decay constant of the oscillation envelope. A relaxation time of 137 nsec was determined, which is in fair agreement with the collisionally induced, homogeneous relaxation time of 112 nsec estimated from collisional linewidth data (see Section 3). This procedure could be a valuable method of rotational relaxation time measurements in HF or other gases.

4.4 CONCLUSIONS AND SUGGESTIONS FOR FURTHER STUDY

Self-induced transparency of HF laser radiation at λ 2.7μ has been observed in low pressure HF gas for several $P_1(J)$ transitions. Low-loss propagation through the resonantly absorbing medium was observed, and intensity threshold values for the onset of SIT were found to agree with those predicted by theory.

The optical nutation effect was also observed in HF gas for strong laser pulses of long duration. The oscillation frequency of the optical nutation was in good agreement with that predicted by theory, and the damping of this oscillation provided a collisionally induced rotational relaxation time in agreement with estimates obtained from collisional linewidth data.

4.4.1 SELF-INDUCED TRANSPARENCY THROUGH WATER VAPOR AND LOSSLESS PROPAGATION THROUGH THE ATMOSPHERE

The HF chemical laser with its high electrical efficiency, high gain, and high, scalable power output is an attractive source of infrared laser radiation. A principal disadvantage is the high absorption of λ 2.7μ radiation by the atmosphere caused by the presence of water vapor.

Self-induced transparency of HF laser radiation resulting from resonance absorption by HF gas would lead one to expect similar effects in resonance absorption by water vapor at low pressures. It is possible that SIT may be observed at higher or atmospheric pressures if pulses are sufficiently short that relaxation is not destructive. A mode-locked HF laser with subnanosecond pulses would be a likely radiation source. Thus SIT could be a valuable means of low-loss atmospheric propagation of HF laser radiation through the atmosphere and is worthy of further study.

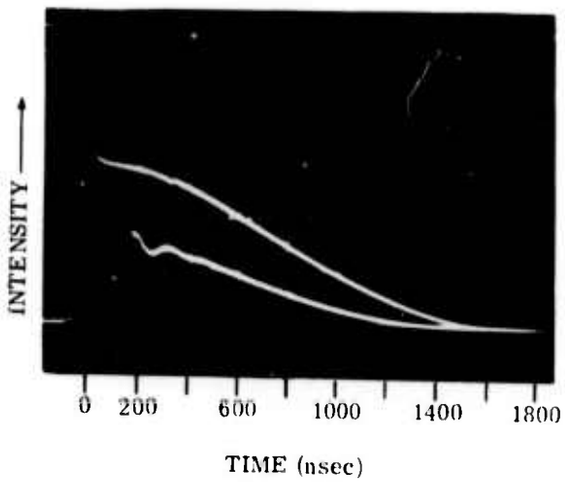


FIGURE 13. OPTICAL NUTATIONS OF HF GAS. The smooth incident $P_1(5)$ HF laser pulse exhibits a 7 MHz oscillation in intensity after passing through 113 mTorr of HF ($\alpha L = 59$). The τp relaxation of the nutation is 15 μsec-mTorr. Each trace is a superposition of about 10 pulses.

4.4.2 PULSE COMPRESSION FOR ATTAINING HIGH PEAK POWER

Using HF gas as the resonant absorber, it is quite possible that HF laser pulses could be compressed in time and enhanced in peak power (see Fig. 10). By focusing an HF laser 3π-pulse through a resonant HF medium to provide appropriate increase in radiation density as the pulse propagates, a long, energetic, HF laser pulse could be compressed into a short, powerful pulse [17]. The power density of present high-power pulsed HF lasers would require the use of a beam expander to decrease the laser E-field so that 3π-pulses could then be achieved for pulse compression. The focusing required for pulse compression would restore the original power density, and the nonlinear propagation effects of SIT would further enhance the power density and shorten the pulse length. Also, if the output of a continuous, HF laser could be mode-locked, the pulse train may be suitable for pulse compression (possibly at high gas pressures).

Pulse compression caused by SIT of water vapor could be valuable for atmospheric propagation. Conceivably, an HF laser 3π-pulse focused so that it has the proper convergence through the atmosphere could undergo pulse compression and increases in peak power as it propagates. The absorption of HF laser radiation by the atmosphere because of the presence of water vapor may enhance propagation via self-induced transparency rather than hinder it.

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